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ELECTRICAL RESISTIVITY, MAGNETIC SUSCEPTIBILITY, THERMOELECTRIC
POWER AND HEAT CAPACITY OF PtGa_2

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**Electrical Resistivity, Magnetic Susceptibility, Thermoelectric Power,
and Specific Heat of PtGa₂**

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The electrical resistivity (ρ), magnetic susceptibility (χ), thermoelectric power (S), and specific heat (C_p) of PtGa₂ were measured as a function of temperature (T). The metallic behavior of this inter-metallic compound is shown from the room temperature resistivity value ($19\mu\Omega\text{-cm}$) and the linear dependence of the S vs. T curve at temperatures above the Debye temperature (θ_D). The diamagnetic susceptibility is independent of T . The density of states (DOS) at the Fermi energy (E_F) obtained from χ and S data agree within 22% and 15%, respectively, of the value obtained previously from a semiempirical band structure calculation. The low temperature C_p data, however, yielded a much smaller DOS at E_F . The data for PtGa₂ are compared to those for Au and AuGa₂ in order to better understand the transport properties of this material.

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I. Introduction

At temperatures above 460K, the intermetallic compound PtGa_2 forms a pseudobinary system with GaAs, whereas elemental Pt reacts chemically with GaAs to form more stable product compounds.¹ Although PtGa_2 is supposed to be a high temperature phase and only a metastable species at room temperature, it is actually quite robust. Single crystals and thin films of PtGa_2 can be grown and examined over the course of years without perceivable disproportionation. Therefore, the study of the electronic and magnetic properties of PtGa_2 is important for understanding its behavior as a potential conducting contact or an active component in optoelectronic circuitry.

PtGa_2 has the cubic fluorite structure, and is isostructural with AuX_2 ($\text{X}=\text{Al}, \text{Ga}, \text{In}$). Jan and Pearson² have reported that AuGa_2 is anomalous in the sense that its thermopower is negative at "low" and "high" temperatures while AuAl_2 and AuIn_2 have positive thermopowers in the temperature range measured (2 to 300K). The ^{71}Ga Knight shift and the magnetic susceptibility of AuGa_2 are strongly temperature-dependent in comparison to its Al and In analogues.³ On the other hand, resistivity^{2,4} and specific heat⁵ measurements display no anomalous variation with temperature in AuGa_2 . In Switendick and Narath's nonrelativistic augmented plane wave (APW) band structure calculation,⁶ a flat band (Γ_2-X_3) lies about 1eV below E_F in AuGa_2 , while for AuAl_2 and AuIn_2 this band disperses strongly and crosses E_F . Kim et al.,⁷ who included the spin-orbit interaction in their mixed-basis band structure interpolation scheme (MBBIS) calculation, reproduced this result. It is generally believed that this flat Δ_2 band, derived from Ga 4s-like anti-bonding states, is responsible for the AuGa_2 anomalies discussed above. However, the observation that the magnetic susceptibility of AuGa_2 between 4.2 and 300K shows a decreasing diamagnetism with decreasing T is still an unresolved issue.⁸ In an angle-resolved photoemission spectroscopy (ARPES) study of AuGa_2 ,⁹ no peak was observed corresponding to the Δ_2 band, although such a flat band should yield an extremely high density of initial states to be sampled.



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The MBBSIS was recently utilized to obtain a semiempirical band structure of PtGa_2 .¹⁰ The flat Δ_2 band was also present in this semiempirical band structure, because the AuGa_2 parameters were used as the starting point in the fit of the DOS to an X-ray photoemission spectrum of the PtGa_2 valence band. However, the ^{71}Ga Knight shift of PtGa_2 is positive and temperature independent, and the conductivity exhibits no anomalous behavior between 4.2 and 300K.¹¹ Since there is no first-principles band structure calculation and very little experimental data published for PtGa_2 , the present study was initiated to provide more information about this potentially interesting material. Section II of this paper describes the experimental procedure. In Sec. III, the results are presented and discussed, and Sec. IV concludes this paper.

II. Experimental procedure

Samples used for the susceptibility measurements were small pieces, with a total weight of 115.4mg, crushed from a PtGa_2 single crystal.¹² A Faraday method, utilizing a Cahn balance, was used for the static magnetic susceptibility measurement in a field of 9 kOe. Temperatures from 4.2 to 300K were measured with calibrated carbon-glass and platinum resistors. In order to verify that the observed magnetization was linear in magnetic field, the susceptibility was measured at several field values at room temperature, liquid nitrogen temperature and liquid helium temperature. The uncertainty in χ is less than 1%.

For the electrical resistivity and thermoelectric power measurements, the same single crystal was cut with a wire saw into a long slice of roughly $10 \times 1 \times 0.5 \text{ mm}^3$ in size. It was then polished with 5 micron diamond grit and cleaned with acetone just before loading into the dewar. The electrical resistivity was measured with a four-probe method. The thermopower was measured between 4.2 and 300K by establishing a temperature gradient across the sample and measuring the voltage developed against Au leads. The Seebeck coefficient (S) was obtained from the slope of a linear least-squares fit of a series of 30 Seebeck voltage vs. thermal gradient measurements. The absolute Seebeck coefficient was derived after subtracting out the contribution of the Au lead wires from the resultant slope. The uncertainty in S is less than 2.5%.

For the specific heat measurements, a PtGa₂ single crystal was cut to about 10×5×1 mm³ in size and polished with 5 micron alumina grit. The weight of the sample was 773.9mg. The ripple method¹³ used in the heat capacity measurements will be described elsewhere.

III. Results and Discussion

A. Electrical Resistivity

Fig. 1 shows the electrical resistivity of PtGa₂; there is no anomaly in the ρ vs. T curve, in agreement with the observation reported in Ref. 11. The room temperature resistivity of PtGa₂, which is only about eight times larger than that of Au, is compared with those of AuGa₂ and Au in Table 1. Compared with the room temperature electrical resistivity values of WSi₂ (35-60 $\mu\Omega$ -cm)¹⁴ and TaSi₂ (40 $\mu\Omega$ -cm)¹⁵, which have been suggested as high temperature non-reactive contacts on GaAs, PtGa₂ is a rather good metal and perhaps to be preferred as a contact for devices. However, the residual resistivity ratio ($\rho_{297.5K}/\rho_{4.2K}$) is only 3.34, which indicates that there are impurities or vacancies in the material. These impurities may also be responsible for the low temperature behavior of S and C_p , as will be discussed in parts (C) and (D) of this section.

B. Magnetic Susceptibility

The measured magnetic susceptibility at 9 kOe is shown in Fig. 2. For PtGa₂, χ has two contributions: one is the temperature-independent diamagnetic susceptibility from the Pt- and the Ga-ion core electrons (χ_i^{Pt} and χ_i^{Ga}), and the other is the conduction electron susceptibility (χ_e). The expression for χ_e also has two components: one is the paramagnetic Pauli susceptibility (χ_e^p), and the other is the Landau-Peierls diamagnetic susceptibility (χ_e^d). For noninteracting free electrons at OK, χ_e^p and χ_e^d are given by

$$\chi_e^p = \mu_B^2 n(E_F) \quad (1)$$

and

$$\chi_e^d = -\frac{1}{3} \mu_B^2 \left(\frac{m_0}{m^*} \right)^2 n(E_F) \quad (2)$$

where μ_B is the Bohr magneton, $n(E_F)$ is the DOS at E_F for both spin directions, and m^*/m_0 is the effective mass ratio. The estimated values for χ_i^{Pt} and χ_i^{Ga} are $-28 \mu\text{emu/mole}^{16}$ and $-9.54 \mu\text{emu/mole}^{17}$ respectively. The net ionic diamagnetism for PtGa_2 is therefore $-47.08 \mu\text{emu/mole}$. Taking $m^* = m_0$ in Eq. (2) and extrapolating χ to OK, we get $n(E_F) = 1.40$ electrons of both spin directions/eV-unit cell, which is 22% larger than the value calculated by the MBBSIS¹⁰. In general, electron-electron interactions lead to an enhancement of the Pauli term by a factor $(1 - \alpha)^{-1}$, where α is the Stoner enhancement parameter. Using the $n(E_F)$ value from the MBBSIS calculation in the Pauli and the Landau-Peierls terms, we estimate α to be around 0.16. Since α usually lies in the range of 0.1 to 0.5,¹⁸ this means that electron-electron interactions in PtGa_2 are very weak. This in turn justifies the use of the free electron approximation in the above calculation of $n(E_F)$.

C. Thermoelectric Power

The S vs. T curve of PtGa_2 is shown in Fig. 3 along with those of Au and of AuGa_2 . Below 12K S of PtGa_2 becomes negative. This can be attributed to trace magnetic impurity scattering, which has also been observed in Au.¹⁹ The shape of the S vs. T curve of PtGa_2 is very similar to that of Au, although their magnitudes differ. This suggests that there may be some similarities in their conduction mechanisms and topology of their Fermi surfaces. As has already been pointed out,¹⁰ PtGa_2 has an Au-like DOS, which explains the gold color of this inter-metallic compound.

Since S of PtGa_2 remained positive at the highest temperature measured, the electrical conduction is by holes.²⁰ This behavior is different from that of AuGa_2 . The calculated flat Δ_2 band of PtGa_2 , which is located within 0.1eV of E_F in the Γ -X direction in the MBBSIS, may actually be either above E_F or may disperse more strongly and cross E_F , as in the case of AuAl_2 and AuIn_2 . The Knight shift¹¹ and magnetic susceptibility data support this suggestion, but to be sure about this point a high resolution ARPES study should be performed.

The occurrence of a maximum in Fig. 3 for the thermopower of PtGa_2 is attributed to the phonon-drag effect.¹⁹ The contribution of this electron-phonon scattering process to S being

positive implies a dominance of Umklapp over normal processes. For PtGa₂, the temperature of this maximum (T_{\max}) is 37.4K, and therefore θ_D is estimated to be $5T_{\max}=187\text{K}$.²¹ The values of θ_D for Au, AuGa₂, and PtGa₂ determined by various methods are presented in Table 1.

For $T \geq \theta_D$, impurity scattering is negligible compared with thermal scattering and the phonon-drag contribution to S is rather small. Hence, diffusion thermopower (S_d) dominates. For metallic conduction, S_d varies linearly with T ,²² and the free electron expression is:²³

$$S_d = \left(\frac{\pi^2 k_B^2 n(E_F)}{3Ne} \right) T, \quad (3)$$

where N is the number of electrons per unit cell, e is the electron charge, k_B is the Boltzmann constant, and $n(E_F)$ is the DOS at E_F .

The dashed line in Fig. 3, which has a slope $(1.035 \pm 0.083) \times 10^{-8} \text{ V/K}^2$, is the least-squares fit to the data points for $T > 187\text{K}$ for PtGa₂. This linear dependence of S with T shows that PtGa₂ is metallic for $T > \theta_D$. Comparing with Eq.(3) and using $N=3$, one may determine that $n(E_F) = 1.27$ electrons of both spin directions/eV-unit cell, which is presented in Table 1 along with those of Au and AuGa₂. The $N=3$ configuration has been used and justified in certain superconducting compounds containing Ga.^{24,25} Pauling²⁶ assigned effective metallic valences of 6 and 3.5 for Pt and Ga, respectively, when they are bonded in intermetallic compounds. The total number of electrons in one unit cell of PtGa₂ is 16 (10 from Pt and 3 from each Ga), and, from simple addition of valence, 13 of them are used to form the Pt-Ga bonds. Therefore, the number of free electrons in one unit cell of PtGa₂ is 3. This explains qualitatively the assignment of $N=3$.

D. Specific Heat

Displayed in Fig. 4 are C_p vs. T data for PtGa₂ for $0.46\text{K} \leq T \leq 4.21\text{K}$ and, in the inset, C_p/T vs. T^2 for $2.34\text{K} \leq T \leq 4.21\text{K}$. At low temperature the specific heat of a metal is represented by an equation of the form:

$$C_p(T) = \gamma T + \beta T^3, \quad (4)$$

where γ and β are the coefficients for the electronic and lattice contributions, respectively, to C_p . If we neglect electron-phonon enhancement, then $n(E_F) = 3\gamma/k_B^2\pi^2$ and $\theta_D = (12\pi^4 k_B N_A N_u / 5\beta)^{1/3}$, where N_A is Avogadro's number and N_u is the number of atoms in a formula unit ($N_u = 3$ for PtGa_2). The data points in the inset of Fig. 4 have been fitted with Eq. (4), yielding the values of $n(E_F) = 0.32$ states/eV-unit cell and $\theta_D = 402\text{K}$. This $n(E_F)$ value is much smaller than the values obtained from the band structure calculation, the magnetic susceptibility, and the thermopower measurements. This apparent smearing of the DOS at E_F may be caused by a temperature-dependent DOS at E_F or/and a contribution arising from magnetic impurities in PtGa_2 . The latter possibility is supported by the resistivity and the thermopower data presented in this paper, while the former is still an open question.

The lattice constants of Au, AuGa_2 , and PtGa_2 are 4.08 Å, 6.06 Å, and 5.911 Å, respectively.^{9,10} Thus, the Brillouin zones for AuGa_2 and PtGa_2 have a smaller volume in reciprocal space than for Au. The $n(E_F)$ value of PtGa_2 should be closer to that of AuGa_2 and higher than that of Au, which is confirmed by the χ and S measurements. Rayne⁵ reported the specific heat data of AuGa_2 between 1.4K and 4.2K and found a considerable curvature in the C_p/T vs. T^2 curve, which, he presumed, is a result of the low θ_D and an anomalously high phonon dispersion for the fluorite structure. In order to take into account this curvature, he added a T^5 term in the expression for $C_p(T)$, which comes from the second term in the expansion of the phonon spectrum. We observed an upturn at $T \cong 2\text{K}$ in PtGa_2 , which is similar to that found in AuGa_2 by Rayne.⁵ By fitting Eq. (4), with the addition of a T^5 term, to PtGa_2 data down to $T = 1.4\text{K}$, we determine $n(E_F) = 0.51$ states/eV-unit cell and $\theta_D = 108\text{K}$. These values are the highest and the lowest obtained for $n(E_F)$ and θ_D , respectively, from the C_p data for PtGa_2 . Thus, although in general C_p data yield the most reliable values for $n(E_F)$ of metals, for PtGa_2 the low temperature anomaly that appears in the C_p vs. T data causes the determination of $n(E_F)$ to be unreliable. Therefore, we have omitted this determination from Table I.

V. Conclusion

No anomalous behavior was found in the electrical resistivity, the magnetic susceptibility, or the thermoelectric power of PtGa_2 . It has a temperature-independent diamagnetic susceptibility. Neglecting electron-electron interaction and using the free electron approximation, the DOS at E_F was determined to be 1.40 electrons/eV-unit cell from the χ data, which agrees with the previously calculated MBBSIS value to within 22%. The S of PtGa_2 resembles that of Au but not that of AuGa_2 . This suggests that the conduction mechanisms and Fermi surface topology of PtGa_2 are similar to those of Au. PtGa_2 has metallic behavior and the conduction is by holes. The flat Δ_2 band appearing in AuGa_2 may be above E_F or may disperse more strongly and cross E_F in PtGa_2 . Assuming free electron conduction and three conduction electrons per unit cell, the DOS at E_F calculated from the S data is 1.27 electrons/eV-unit cell, which agrees with the value calculated with the MBBSIS to within 15%.

The specific heat of PtGa_2 behaves anomalously below $T \cong 2\text{K}$, which may be the result of a temperature-dependent DOS at E_F or/and a contribution arising from magnetic impurities. The DOS at E_F obtained from the C_p data is much smaller than the value calculated from the MBBSIS or the values obtained from χ and S measurements.

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Table I. Comparison of values of ρ , θ_D and $n(E_F)^a$ for Au, AuGa₂ and PtGa₂.

	ρ ($\mu\Omega$ -cm) at 20°C	θ_D (K)	$n(E_F)$
Au	2.24 ^b	165 ^c	0.31 ^d
		162.4 ^e	0.18 ^f
		161.6 ^g	0.24 ^h
AuGa ₂	12.9 ⁱ	245 ^j	1.12 ^k
		235 ^c	1.14 ^m
		192 ^m	
PtGa ₂	19.05 ⁿ	187 ^c	1.09 ^f
			1.27 ^o
			1.40 ^p

- a. DOS at E_F in units of number of electrons per eV per unit cell for both spin directions.
- b. Handbook of Chemistry and Physics (College Edition), (Chem Rubber Co.: 1984), p. F-120.
- c. Estimated from thermoelectric power measurements, $\theta_D \cong 5 T_{\max}$.
- d. Calculated from D. L. Martin's specific heat data (Phys. Rev. **141**, 141 (1966)) using $n(E_F) = 3 \gamma / k_B^2 \pi^2$, where γ is the intercept of the C_p/T vs. T curve.
- e. From specific heat measurement, D. L. Martin, Phys. Rev. **141**, 141 (1966).
- f. From MBBSIS band structure calculation, Ref. 10.
- g. From zero-temperature elastic constants measurement, G. A. Alers, in *Physical Acoustics*, W. P. Mason, ed. (Academic Press Inc., New York), Vol. IIIB, Chap. I.
- h. Calculated from thermoelectric power data, Ref. 16, at $T > \theta_D$ using Eq. (3) in this paper and $N=1$.
- i. From resistivity measurement, Ref. 2.
- j. Calculated in Ref. 5 from the resistivity data of Ref. 2.
- k. From MBBSIS band structure calculation, Ref. 7.
- m. From specific heat measurement, Ref. 5.
- n. From resistivity measurement, this work.
- o. From thermoelectric power measurement, this work.
- p. From magnetic susceptibility measurement, this work.

FIGURE CAPTIONS

- Fig. 1.** Electrical resistivity of PtGa_2 as a function of temperature from 4.2 to 300K.
- Fig. 2.** Magnetic susceptibility of PtGa_2 and AuGa_2 as a function of temperature from 4.2 to 300K. The AuGa_2 data were taken from Ref. 3.
- Fig. 3.** Temperature dependence of the thermoelectric power $S(T)$ for Au, AuGa_2 , and PtGa_2 . The data for Au and AuGa_2 are taken from Ref. 12 and Ref. 2, respectively. The dashed line is the least-squares fit to PtGa_2 data for $T > 187\text{K}$.
- Fig. 4.** Specific heat C_p vs. temperature T for PtGa_2 for $0.46\text{K} \leq T \leq 4.21\text{K}$. Inset: C_p/T vs. T^2 for $2.34\text{K} \leq T \leq 4.21\text{K}$. The solid line represents a least-squares fit of Eq. (4) in the text to the data.

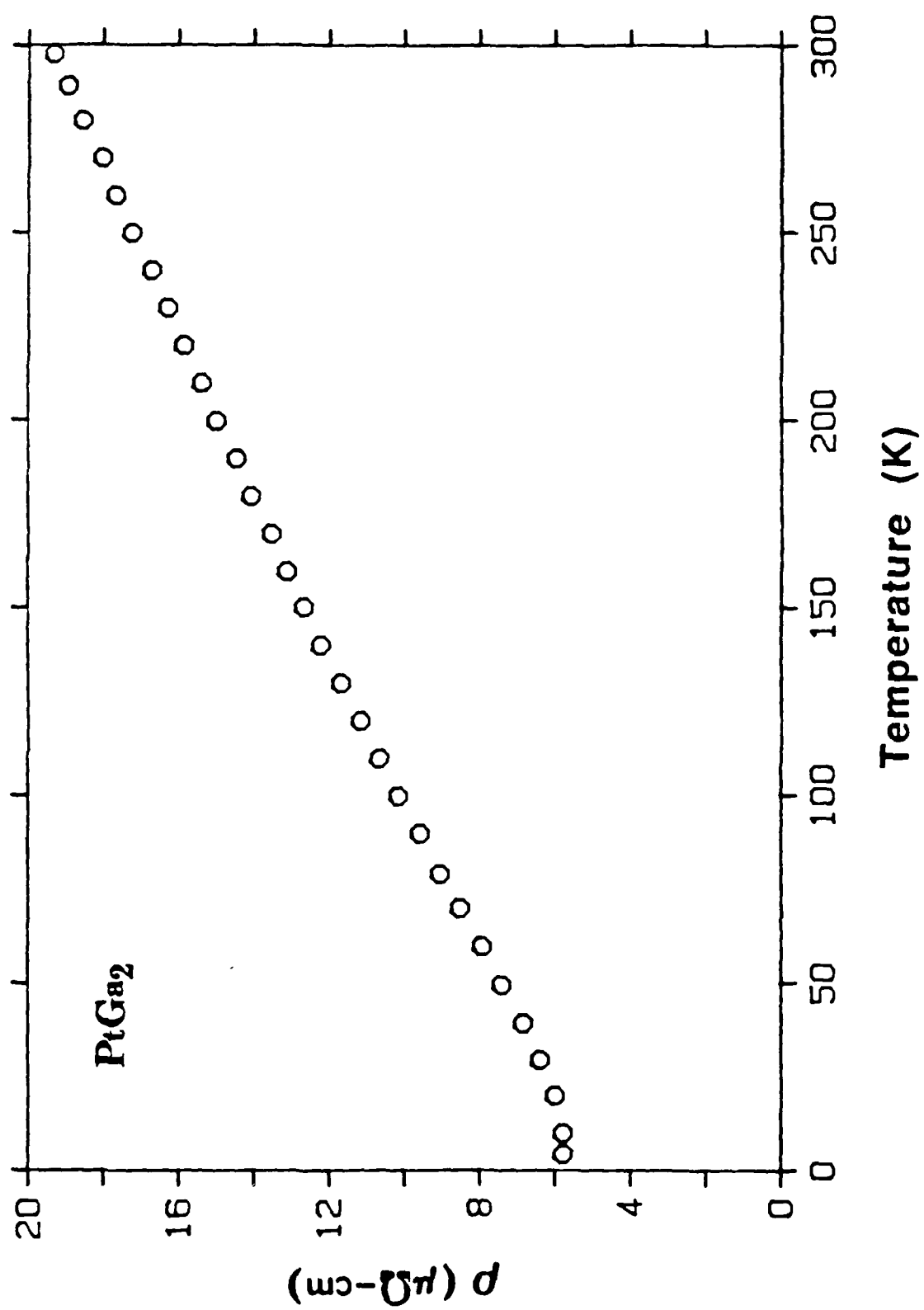


FIG. 1

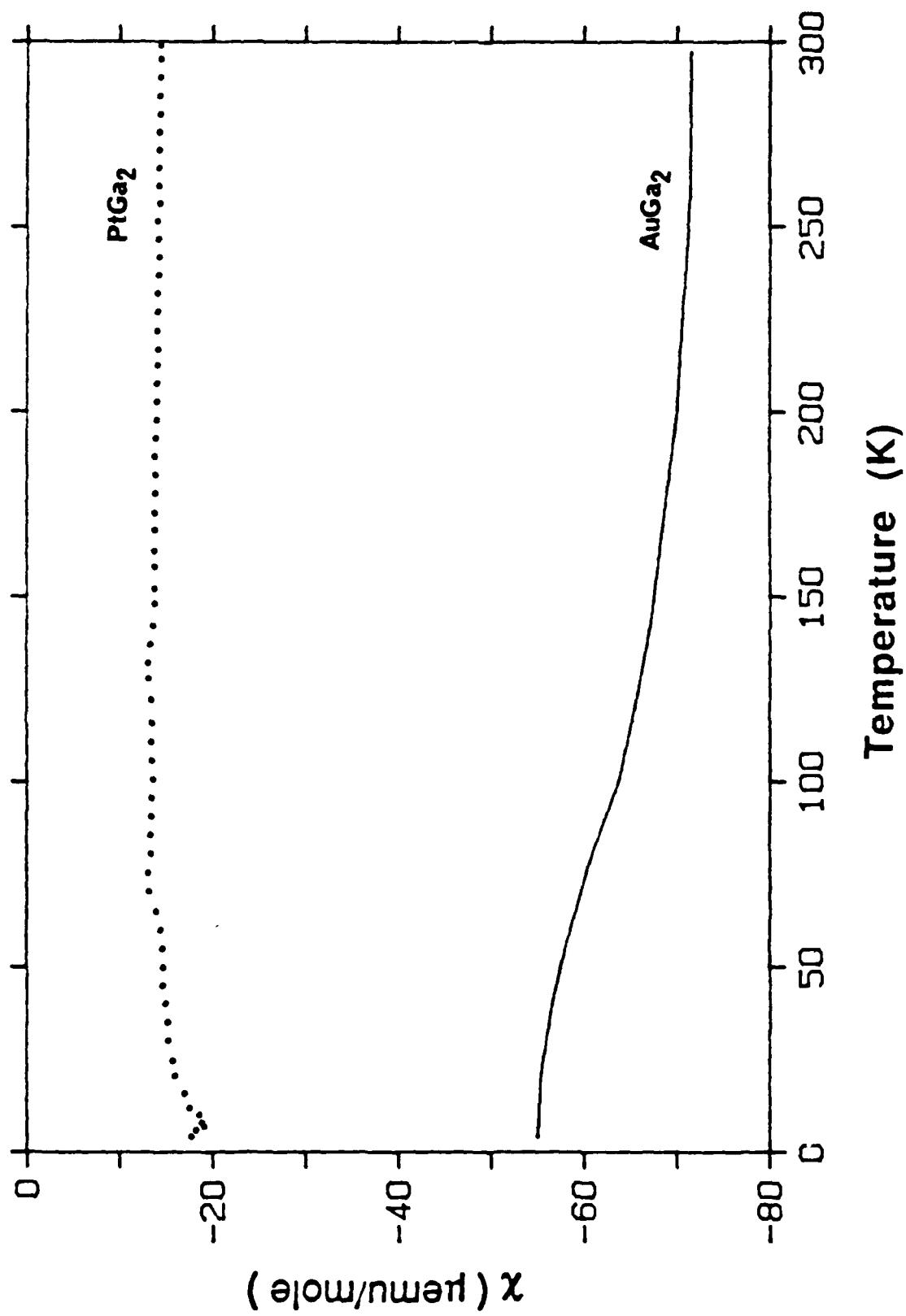


FIG. 2

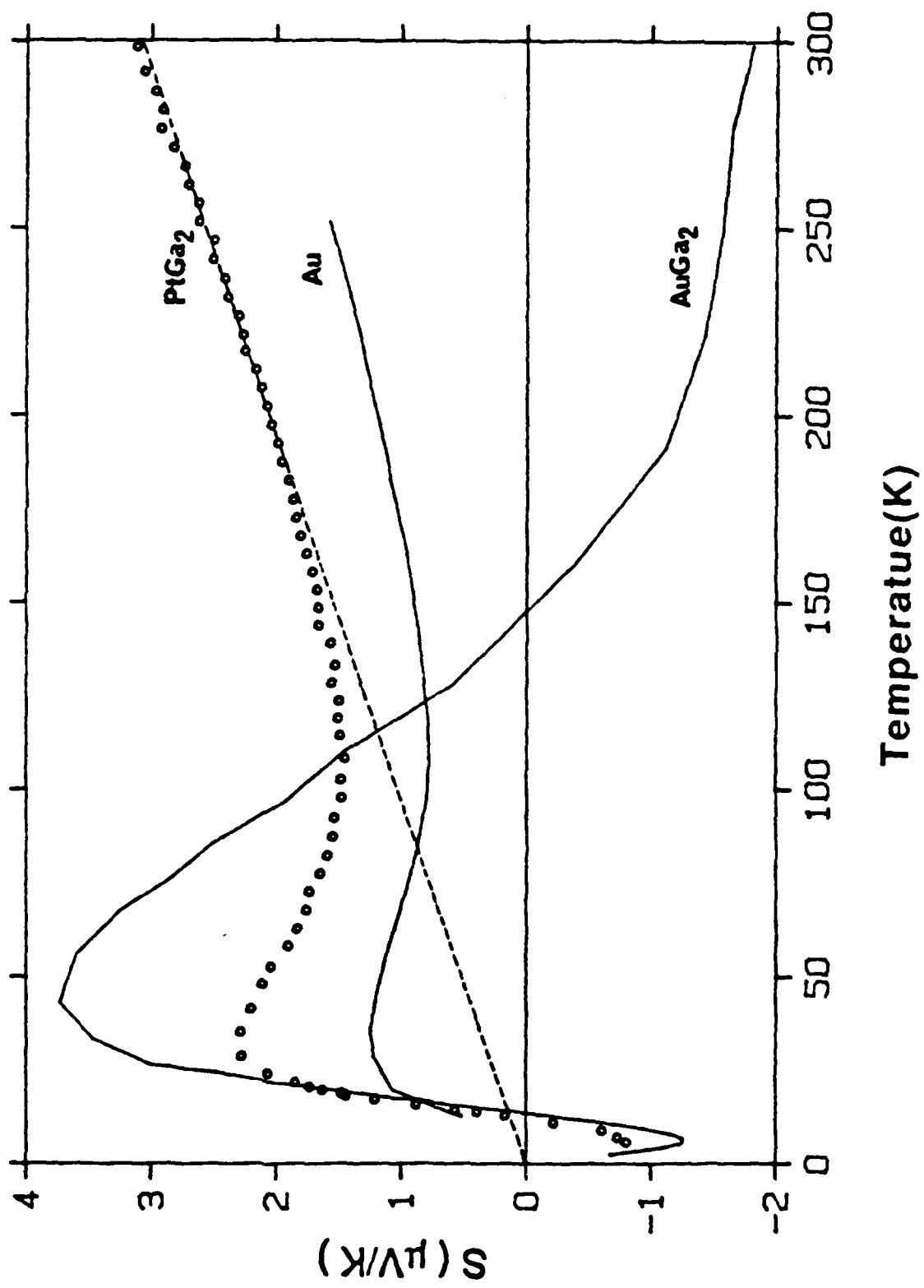


FIG. 3

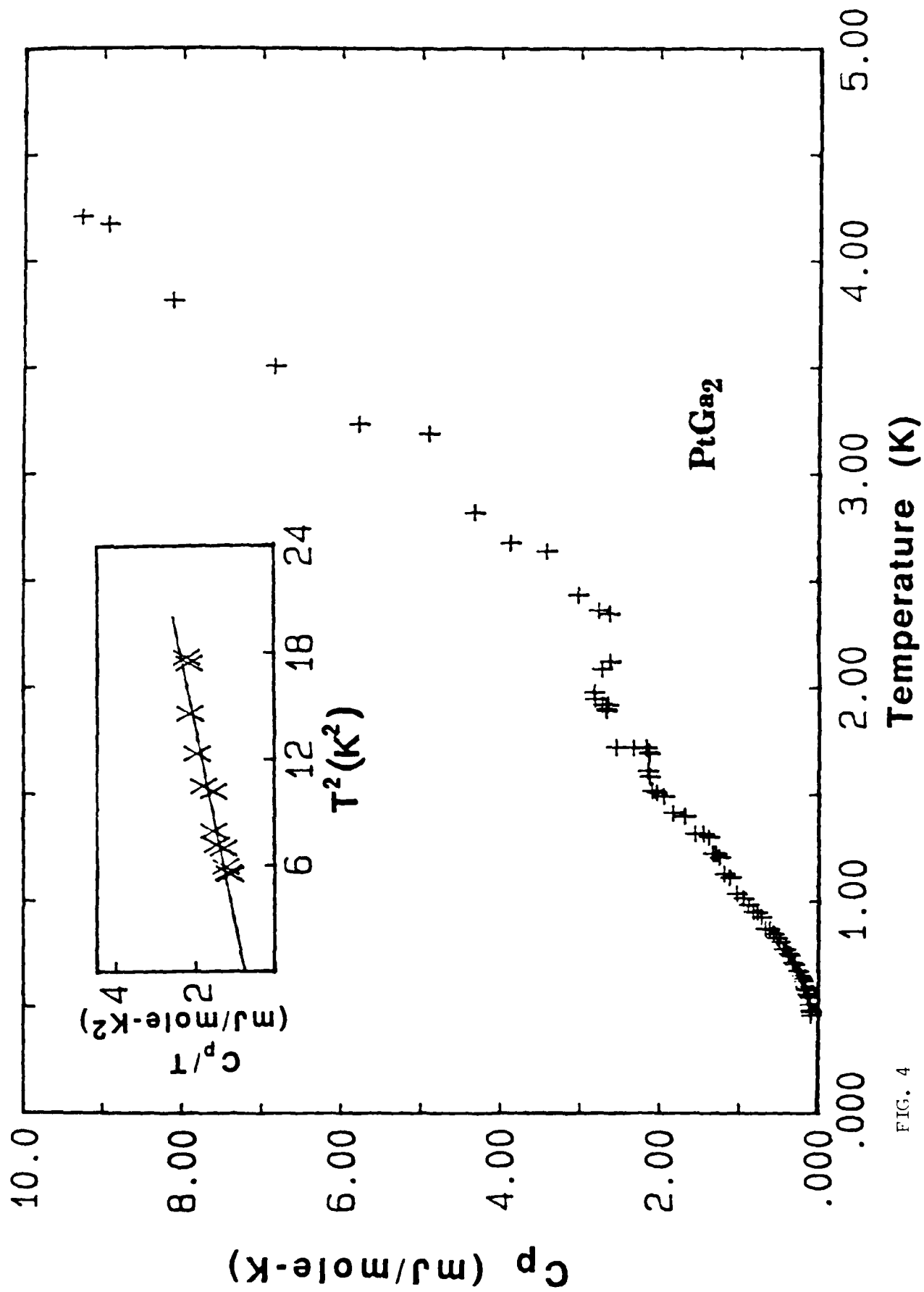


FIG. 4